

Interactive comment on “Organic molecular composition of marine aerosols over the Arctic Ocean in summer: contributions of primary emission and secondary aerosol formation” by P. Q. Fu et al.

Anonymous Referee #1

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This manuscript presents new chemical data obtained from aerosols collected over the Arctic Ocean during summer. The manuscript is significant since it presents a new data set that has been lacking in the literature, especially for the Arctic troposphere. This manuscript should certainly be published in Biogeosciences as a discussion paper. The authors should know that I have already carefully read through this paper and have several comments that I would like to make now, all of which I hope are constructive and useful in terms of improving the quality of the manuscript. The current quality of the manuscript is quite good; however, my comments will hopefully be useful in presenting

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these new exciting data in the clearest way possible. In order to jump start the review process, I will post my major comments below so that the authors can consider them during the discussion phase.

Major Comments/Suggestions:

- 1.) The major weakness of this study is that the authors could be missing important organic species that are not amenable to GC/MS analyses. By not measuring other tracer compounds, such as organosulfates that are easily observable by LC/ESI-MS techniques, the authors could be missing important sources or atmospheric processes. As clearly noted in a recent review article by Hallquist et al. (2009, ACP), in order to capture the organic aerosol formation well, several advanced analytical techniques should be used in tandem, as many individual techniques (such as GC/MS) have their respective weaknesses. For example, it is known with GC/MS that one can not characterize more than 10-20% of the organic aerosol mass at the molecular level.
- 2.) Another issue I have with this study, which will not prevent this study from being published, is why wasn't a PM_{2.5} high-volume sampler used? This would help to isolate the collection of climatically relevant size ranges. With TSP you are getting everything that is present in the atmosphere at that time. As I'm sure the authors know, PM_{2.5} high-volume samplers certainly exist and would have been preferred here in the design of this study, especially if you argue the research you are doing is relevant to climate change, etc.
- 3.) Why didn't the authors consider filtering solvent extracts through a Teflon membrane syringe filter instead of with their quartz wool packed in a Pasteur pipette approach? I worry the quartz wool might cause permanent absorption (and thus losses) of some organic constituents. Have the authors tried using authentic and surrogate standards to test for the recovery of compounds through this quartz wool?
- 4.) In section 3.1, it might be helpful to add a table (or to an existing table) that summarizes the origins of air masses for each filter collected. This might be easier for readers

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to find this information without having to search the manuscript for these details.

5.) Based on what is now known about isoprene SOA formation, I wonder if the authors have particulate sulfate (as well as particle-phase inorganic) data available from their campaign? Did anyone from your campaign make ion chromatography (IC) measurements from the aerosol samples? If so, it would be very interesting to know if the 2-methyltetrols and C5-alkene triols correlate with sulfate mass loadings.

Further, do you or your colleagues from the campaign have gaseous chemical data? Specifically, it would be interesting to know if 2-methylglyceric acid correlates with NO_x.

Lastly, since you are using GC/MS, do you measure the other IEPOX-derived SOA tracers? Specifically, I mean do you measure cis- and trans-3-methyltetrahydrofuran-3,4-diols (see recent paper by Lin et al., 2012, ES&T). I would like to further note that since you see 2-methyltetrols and C5-alkene triols, you likely will observe the organosulfates derived from IEPOX if using ESI-MS techniques (operated in the negative ion mode). The organosulfate of IEPOX is an important compound since a recent study by Froyd et al. (2010, PNAS) showed that this compound is one of the single most abundant compounds in the free troposphere. This could indicate that it has a long atmospheric lifetime, allowing it to be transported long distances. The Froyd et al. (2010, PNAS) study used on-line aerosol mass spectrometry data collected from several aircraft campaigns. I should note that their aerosol mass spectrometer is equipped with laser ionization.

7.) The authors seem to indicate that the ratios of 2-methylerythritol to 2-methylthreitol are lower than what has been seen before, especially at lower latitudes. They suggest that this could indicate that some of the 2-methyltetrol contribution is derived from primary biogenic emissions, as recently proposed by Noziere et al. (2011). However, could this be due to one isomeric epoxide produced in the gas-phase oxidation of isoprene (Paulot et al., 2009, Science) leading to this difference?

8.) The authors discuss that the C5-alkene triol to 2-methyltetrol ratio was high on one

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foggy (humid) day and lower on another foggy (humid) day. If the authors had IC data, they could see if this is due to sulfate loading. Furthermore, since aerosol acidity has been previously shown by Surratt et al. (2010, PNAS) and Lin et al. (2012, ES&T) to be important in promoting the heterogeneous chemistry of gaseous IEPOX, have the authors also considered trying to estimate aerosol acidity from IC data (if available)?

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